Open-loop recycling of PET-PE post-industrial multi-layered plastic waste

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ABSTRACT: In our present society it is difficult to imagine life without high-performance plastic packaging materials. Apparently 'simple' trays and films for packaging of all kinds of (food) products often consist of several layers of different polymer materials, each of which contributes its own functionality to the packaging. However, the various layers are joined together physically so that the constituent polymers can no longer be separated from one another after use and during subsequent recycling they must be processed together as a mixture. This has a considerable effect on the properties and processability of these materials, which makes efficient mechanical recycling very challenging. Therefore so far much of this waste goes to incineration with energy recovery or ends up in landfills. The aim of this research is to examine the possibility for open loop recycling of a post-industrial multi-layered waste stream and to compare the effect of different compatibilizers. The resulting objective is to identify and develop a suitable application or demonstrator for the multi-layered waste using the principles of 'Design From Recycling', a material-driven design approach. Therefore a multi-layered tray and foil consisting of PET and PE, initially used for sliced meat packaging, is investigated and is first subjected to intensive characterization. Additionally, the waste is melt-blended with different percentages of compatibilizers to improve both mechanical and rheological properties as well as the reprocessability. Afterwards, open-loop recycling into a multifilament extrusion application and a tape extrusion application is evaluated.

1 INTRODUCTION

In multi-layered packaging materials, different polymers are layered next to each other in one foil or sheet to maximize the performance of the packaging material. Each polymer actually contributes his own functionality to the layered packaging material. Polyethylene terephthalate (PET) is often used for his low permeability for different gases and high mechanical strength while polyethylene (PE) is applied because of its reasonable price, excellent sealing properties, water barrier properties and cold temperature performance. Ethylene-vinyl alcohol (EVOH) is often used in a thin layer because of his excellent barrier properties. Common combinations in multi-layered foils and sheets are therefore PET/PE and PET/PE/EVOH/PE. EVOH is used between the PE layers to cover for his poor water resistance (Wagner Jr 2016).

The various layers are joined together physically, so that the constituent polymers can no longer easily be separated from one another after use and during recycling they must be processed together as a mixture. This has a considerable effect on the properties and processability of these materials, which makes efficient mechanical recycling very challenging. Therefore until now much of this waste goes to incineration with energy recovery or ends up in landfills (Ragaert, Delva et al. 2017).

As a result, according to Plastics Recyclers Europe, prior to 2014 annually 700000 tonnes of PET containing food trays were not sorted out and not recycled in Europe (Tarlowska 2014).

In order to be able to use this waste stream for new closed-loop or open-loop products, the mechanical properties must be improved. This can be done either by using additives that improve miscibility or specific custom processing techniques.

The purpose of this study is to examine whether the mechanical properties and the morphology of a recycled post-industrial waste stream consisting of PET trays and their topfoils can be improved by adding different amount of two compatibilizers: LLDPE-g-MA and an ethylene terpolymer. Furthermore, based on the principles of Design From Recycling (Ragaert, Hubo et al. 2017), open-loop recycling into a multifilament extrusion application and a tape extrusion application is evaluated.

2 MATERIALS AND METHODS

2.1 *Materials*

The base material is a mixture of post-industrial trays and their covering foils provided by the Belgian company Colruyt. This mixture consists mainly of PET and PE. The composition of both topfoil and tray is approximately 80/20 vol% PET/PE(+EVOH). This base material is the matrix for four formulations with two different compatibilizers, described in Table 1.

The two additives used are:

1. Compatibilizer 1: Tecnobond cfa/s from Tecnofilm, which is a linear low density polyethylene grafted with maleic anhydride (LLDPE-g-MA;
2. Compatibilizer 2: Elvaloy PTW from DuPont which is an ethylene terpolymer.

Table 1: Nomenclature and composition of the different blends.

Name	PET/PE [wt%]	Additive [%]	Additive
PET/PE	100	-	-
10% comp 1	90	10	LLDPE-g-MA
10% comp 2	80	20	Ethylene terpolymer
20% comp 1	90	10	LLDPE-g-MA,
20% comp 2	80	20	Ethylene terpolymer

2.2 *Sample preparation*

Shredding of the multi-layered material into feedable sizes was done on a Hellweg shredder.

Compounding of small sample amounts (up to 7 g) was done on a ThermoFisher™ HAAKE™ miniCTW. Tensile bars and impact bars were produced using a ThermoFischer™ Minijet Pro.

Upscaling was performed using a Leistritz ZSK 27 twin screw compounder (scale 5-20 kg/h) including melt filtration. Barrel temperatures were set at 230-250-265-270-275-280-285 °C.

The compounded material was further processed in ISO tensile bars using an Arburg 320 S allrounder 500-150 injection moulding machine. Barrel temperatures during the injection moulding were set at 255-260-260-265-265 °C.

2.3 *Characterization*

The mechanical properties of the different materials were evaluated via tensile tests on a ZwickiLine (testControl II) Z2.5 and via Charpy Impact tests according to respectively the ISO 527-2 and ISO 179 procedure.

Scanning Electron Microscopy (SEM) analyses using a JEOL JSM-7600F electron microscope were performed on the middle section of a tensile bar. To isolate this section, a notch was applied on the sample, next, it was frozen under liquid nitrogen and in the last step it was broken. In this way, a smooth fracture surface was obtained that enabled the evaluation via SEM. Prior to image recording, the samples were coated with a nm-thick Pt coating.

3 RESULTS

3.1 *Mechanical properties*

The tensile and impact properties of the PET/PE and the compatibilized samples are presented in Table 2 The PET-PE recycled material has a Young's modulus over 2 GPa, which is relatively high for these types of materials. The impact strength, however, which is a measure for the toughness of the material, is quite low with a value only slightly above 2 kJ/m².

Two different (LLDPE-g-MA and ethylene terpolymer) compatibilizers were added in 10 and 20 wt%. It can be observed that both compatibilizers reduce the Young's modulus and tensile strength of the PET/PE base material. On the other hand, however, strain at break and impact strength are positively influenced by both.

Table 2: Mechanical properties of the different blends.

	Young's modulus ± stdev [MPa]	Tensile strength ± stdev [MPa]	Strain at break ± stdev [%]	Impact strength ± stdev [kJ/m²]
PET/PE	2216 ± 99	43 ± 1	47 ± 33	2,15 ± 0,51
10% comp 1	1832 ± 90	39 ± 1	79 ± 75	2,80 ± 0,24
10% comp 2	1718 ± 49	35 ± 1	75 ± 51	7,34 ± 1,31
20% comp 1	1280 ± 83	32 ± 2	150 ± 51	2,90 ± 0,18
20% comp 2	1176 ± 43	28 ± 1	150 ± 33	9,88 ± 1,80

Comp 1: LLDPE-g-MA; comp 2: ethylene terpolymer

Compatibilization and impact modifying often results in a trade-off relationship between two important parameters i.e. stiffness and impact strength. This important relationship is therefore plotted in Figure 1. It is clear that compatibilizer 2 offers the best combination for impact strength and stiffness. Sample 10% comp 2 increases the impact strength with almost 400% while only reducing the stiffness to 1,8 GPa, which is still an acceptable value for numerous applications. Higher toughness values can be reached by increasing the loading of the compatibilizer to 20 wt%, although this increase reduces the Young's modulus down to 1,4 GPa.

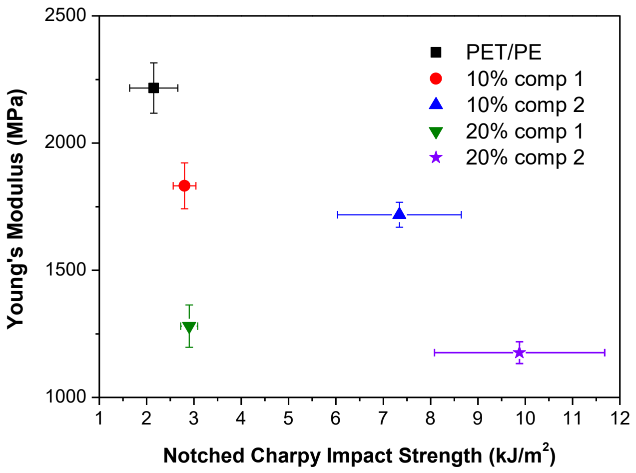

Figure 1. Plot of Young's Modulus versus Notched Impact Strength. Error bars represent one standard deviation.

3.2 *Morphology*

The morphology of the different blends is shown in Figure 2. The PET/PE blend shows a typical dispersed morphology with spherical PE particles with varying diameters between 8 μm and 35 μm distributed poorly in the PET matrix.

The grafted polyolefin does not succeed in reducing the particle size of the dispersed phase. In order to do this, the compatibilizer has to be able to migrate to the interphase and reduce the interfacial tension. This was probably not the case as the maleic anhydride groups are known to be very reactive towards the hydroxyls end groups of PET, which are abundant because PET is the matrix material. This coarse morphology also explains why only a small improvement in impact strength was measured.

It is observed that, coherent with the good impact properties, the dispersion and distribution of the PE particles in the PET matrix is very good in case of compatibilizer 2 resulting in a more homogenous material.

4 PRELIMINARY EVALUATION OF OPEN-LOOP APPLICATIONS

4.1 *Multifilament extrusion*

Preliminary multifilament extrusion trials were performed on a semi-industrial spinning line (Spinmaster). A bundle of filaments was extruded, cooled in air and winded on heated rolls. A stretching rate of 3,3 was applied. A winding speed of 1800 m/min was used.

First experiments with a PET/PE showed that multifilament extrusion as such is not possible due to the incompatible matrix which induces early breakage of the filaments. On the other hand when 5 % of compatibilizer 2 (ethylene terpolymer) is added to a blend of PET and PE multifilament extrusion is feasible and the obtained filaments have mechanical properties slightly lower than virgin PET. Reprocessing of the post-industrial waste considered in this article on the other hand showed some additional problems. Contaminations caused by the presence of paper of meat leftovers are having a significant influence on the processability. Additional melt filtration trials are needed to purify the material.

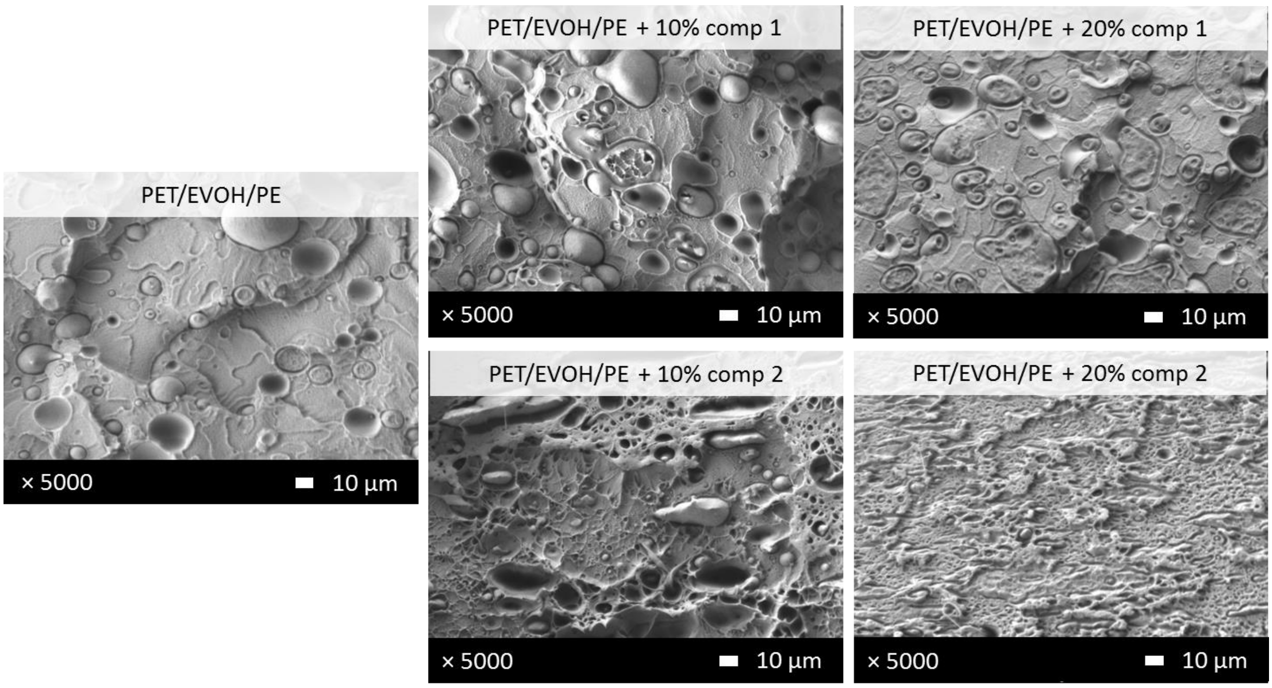

Figure 2. SEM images of the different blends.

4.2 *Tape extrusion*

Tape extrusion trials were performed on a semi-industrial extrusion line consisting out of an extruder, a water bath for quenching, stretching rolls and an oven for additional stretching. A draw ratio of 8 to 10 is aimed for in order to obtain the desired mechanical properties. First promising results are obtained with the post-industrial waste when the compatibilizer 2 (the ethylene terpolymer) is added in 5 wt%. Also in this case further melt filtration is needed since the draw ratios and the mechanical properties are now limited due to the presence of contamination.

5 CONCLUSION

In this paper, a two layered tray and foil consisting of PET and PE, initially used for sliced meat packaging is additivated and mechanically recycled with different compatibilizers. It can be concluded that the ethylene terpolymer is much more effective in compatibilizing both phases then the LLDPE-g-MA. This was attributed to a rubber toughening mechanism for the first compatibilizer, while the LLDPE-g-MA was not able to act and react at the interphase.

The preliminary experiments on multifilament extrusion and tape extrusion showed that reprocessing of PET-PE blends should be feasible by using compatibilizer 2. Further research is needed in order to determine the purification steps and to optimize the most suitable compatibilizer and dosage of compatibilizer making it possible to stretch the materials and to obtain the desired mechanical properties.

6 ACKNOWLEDGEMENTS

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7 REFERENCES

Ragaert, K., L. Delva and K. Van Geem (2017). "Mechanical and chemical recycling of solid plastic waste." Waste Management **69**(Supplement C): 24-58.

Ragaert, K., S. Hubo, L. Delva, L. Veelaert and E. Du Bois (2017). " Upcycling of contaminated post-industrial polypropylene waste: A design from recycling case study." Polymer Engineering & Science **58**(4): 528-534.

Tarlowska, E. (2014). PET trays existential dilemma, Plastics Recyclers Europe.

Wagner Jr, J. R. (2016). Multilayer flexible packaging, William Andrew.